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Time Dependent Synthesis of Cadmium Sulphide Thin Films For Solar Cell Applications

[1]Almas Z.Shaikh, [2]Kakasaheb C. Mohite, [3]Mehboob Nagarbawdi
[1]Department of Physics, Haribhai V. Desai College, Pune, 411002, MS, India
[2]ChandmalTarachand Bora College, Shirur, Pune-412210 , MS, India
[3]ARHAM ,International Insitute of Information Security, Pune. 411001 , MS, India

Abstract : Chemical bath deposition (CBD method was carried out at 60°C for Cadmium Sulphide (CdS) thin films. The optical and structural properties of thin films were systematically investigated under the influence of different deposition time with an interval of half anhourThe UV-Visible spectroscopy study showed high optical transmittance in the visible and NIR region of solar spectrum. The optical band gap varied from 2.4eV to 2.1eV with deposition time. XRD analysis revealed the polycrystalline hexagonal phase of CdS thin films.

Keywords: CdS, CBD, UV-Visible spectroscopy, XRD

1. INTRODUCTION:

CdS is II –VI, n-type binary compound semiconductor material of high optical transmittance, direct wide band gap (2.42eV), high index of refraction[1,2]. CdS has vast applications in photo-conducting cells [2], photo-sensors [3], transistors [4], optical waveguides and non-linear integrated optical devices[5]. In addition, the chalcogenides thin film CdS is most popularly used as n-type window layer in CdTe and CIGS solar cell [6]. There are enormous physical and chemical vapour deposition methods for thin film deposition [7] such as, magnetron sputtering [8], electrodeposition [9], spray pyrolysis [10], successive ionic layer absorption reaction (SILAR) [11], chemical bath deposition [12]. The chemical bath deposition (CBD) method is an attractive technique on account of many features like it is low cost, simple, and non-vacuum based low deposition temperature technique with zero maintenance. CBD is capable to produce uniform, adherent transparent and reproducible layer at mass scale [13].
The objective of the present study is to investigate the structural and optical properties of CdS thin films prepared using CBD method. The Optical properties of thin films were analysed using UV-Visible spectroscopy and structural properties were characterized with X-ray diffractometer (XRD).

2. Methodology:

CBD is a chemical solution growth technique; the chemical bath is prepared using dilute alkaline solution of metal ion and chalocogenide source. Complexing agent assists in releasing and controlling cadmium (Cd^{2+}) and reacts with sulphur (S^{2-}) ions to form CdS molecules [14]. The CdS formation is detailed in the following series of chemical reactions Eq.(1-4)[15].

$$Cd\left((NH_3)\right)_4^{2+} \rightleftharpoons 4\mathrm{NH_3} + \mathrm{Cd}^{2+} \quad (1)$$
$$(NH_2)_2CS + OH^- \rightleftharpoons \mathrm{SH}^- + \mathrm{H_2O} + \mathrm{C(NH)_2} \quad (2)$$
$$\mathrm{SH}^- + OH^- \rightleftharpoons \mathrm{S}^{2-} + \mathrm{H_2O} \quad (3)$$
$$\mathrm{Cd}^{2+} + \mathrm{S}^{2-} \rightleftharpoons CdS \quad (4)$$

Substrate cleaning is the essential step in the formation of pinhole free, homogenous, uniform and adherent thin films using CBD method. Microscopic glass slides of dimension (75×25×1) mm were used as substrate. The slides were cleaned using chromic acid solution followed by the concentrated HCl solution for 10 minutes and finally washed with the distilled water and then dried in air atmosphere [16] .

The chemical solution of 0.012 M cadmium chloride was prepared in 100ml distilled water and 0.03M of NH_4Cl was added into it subsequently. Further, 5 ml ammonia was added drop wise with slow rate till chemical bath become alkaline. The substrates were clamped vertically and fixed in the bath solution. When bath temperature was reached to 60ºC, 0.03M thiourea was added into the solution. The color of solution was changed to dark yellow indicating formation of CdS molecules. The heating was continued till 0.5 hours and then deposited slides were removed from the solution. The deposited slides were rinsed 3 to 5 times with distilled water to remove loosely attached CdS colloids and finally the slides were dried in an open air atmosphere. The deposition process was repeated at different deposition time 1, 1.5, and 2 hours.

UV-Visible spectrophotometer (SHIMADZU UV-1800) of dual beam was used to analyse the optical transmittance in the wavelength range of 200-1000 nm at room temperature and optical band gap of CdS. The structural properties of CdS thin films were investigated using Cu-Kα X-ray with 2θ values ranging from 20 to 80º. The grain size was calculated using Debye Scherer formula.

3. Results and Discussions:

3.1 Optical Study:

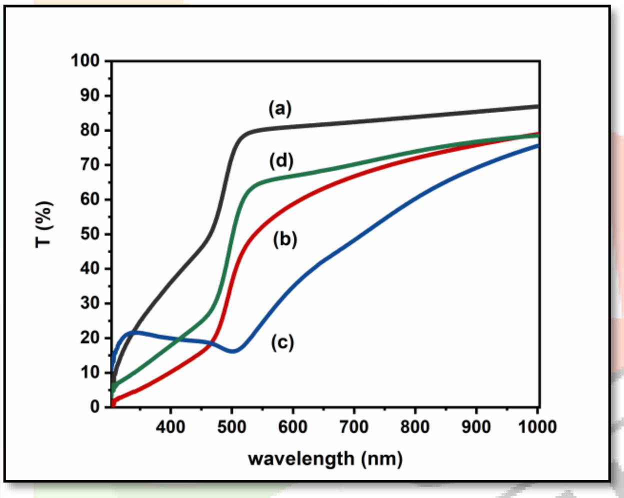

Fig.1 Optical transmittance Spectrum of CdS for different deposition time (a) 0.5, (b)1, (c) 1.5, and (d) 2 hours

Figure 1 displays the optical transmittance spectra of CdS thin films deposited for deposition time from 0.5 to 2 hours with constant time interval of 0.5 hours and Table 1 demonstrates the average transmittance over the wavelength ranges from 500 to 1000nm. The CdS thin film deposited for 0.5 hours showed highest average optical transmittance (83%) among all deposited films.

Thin film deposition by chemical solution illustrates the homogeneous precipitation which results in the formation of colloids and beginning of nucleation and growth of thin film and the heterogeneous growth causes ion by ion condensation of Cd^{2+} and S^{2-} ions to form CdS on the substrate. The homogeneous growth mechanism of CdS on the substrate caused formations of large colloids of CdS molecules and results into decrease in optical transmittance from 83 to 58%[17–19].

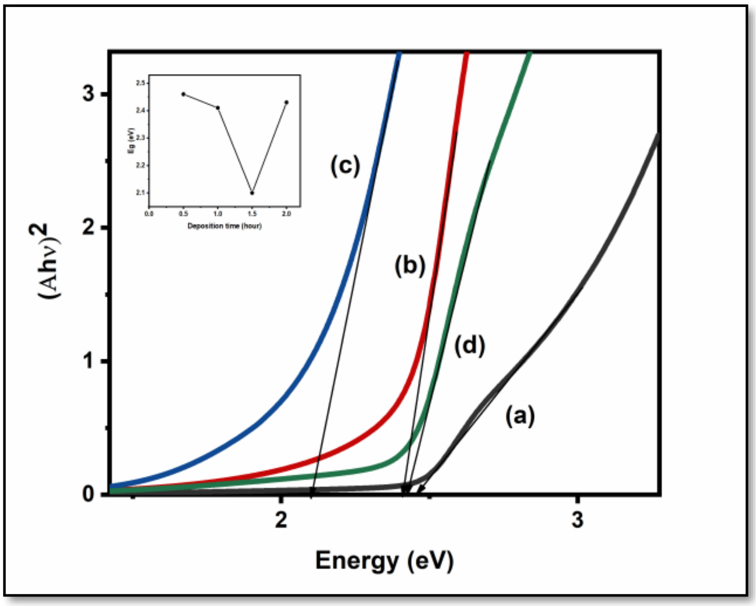

Fig.2. :Tauc plot of CdS for different deposition time (a) 0.5, (b)1 , (c) 1.5, and (d) 2 hours

However, further increasing the deposition time to 2 hours enhanced the transmittance to 71%, and the heterogeneous reaction mechanism is dominant for longer deposition. The UV radiations are easily absorbed by CdS whereas NIR and visible radiations are entirely transmitted by CdS due to higher energy of UV radiations and lower energy of NIR visible radiations than the band gap of CdS respectively. The sharp fall of transmittance is observed at band edge between 450-500nm for all thin films except for the thin film of 1.5 hours deposition. The sharp fall shows formation of smooth and uniform thin film[20]. The transmittance in the low wavelength region extends up to 300 nm indicating the presence of disorders or amorphous components in the film[21]. Figure 2 emphasises the Tauc plot of CdS thin films at different deposition time. It was used to obtain the optical band gap[22] , and the values are listed in the Table 1.

Table 1 Band gap and average Transmission of the CdS thin films prepared at different deposition time

Deposition time (hours)	Average Transmittance (%)	Band gap (eV)
0.5	83	2.46
1	64	2.41
1.5	59	2.1
2	71	2.43

The calculated band gap energy of CdS thin films is found to decrease from 2.46 to 2.1eV when deposition time increased from 0.5 to 1.5 hours respectively and inset of Fig.2 shows the nature of change in band gap with deposition time. This decrease is due to the presence of impurities, grain size, carrier concentration, deviation from stoichiometry of the film and lattice strain[23–25]. These band gap values are comparable to and well aligned with earlier experimental findings[26]. Figure 2 shows that further increase in the deposition time for 2 hours shifted the band edge to lower wavelength and corresponding band gap obtained is 2.43eV[27]. The optical analysis of present work emphasis suitability of the material as a window layer in solar cell applications[28,29].

3.2 Structural Study:

Figure 3 shows the X-ray diffraction pattern of CdS thin films deposited at bath temperature 60˚C for different deposition time 0.5, 1, 1.5, and 2 hours. XRD pattern shows a single sharp and high intense diffraction peak at 2θ= 28.4º, corresponds to reflection plane (101) in all deposited CdS thin films except for the film of 1.5 hours deposition with slight shift in Bragg peak to 28.35º[30-38].

The additional weak XRD peaks at 25.5 and 26.8º with the planes of reflection oriented along (100) and (002) are pointed for CdS thin film deposited for 2 hours respectively. All diffraction peaks were compared with JCPDS card 80-0006 and confirmed the formation of hexagonal phase of CdS as reported in the literature and previous research work[31,32]. Figure 3 replicates significant decrease in FWHM and corresponding increase in the intensity of Bragg peak with deposition time and thus enhanced the crystallinity of the material owing to increase in decomposition of reactants and production of ions with deposition time[33]. The calculated interplanarspacing corresponding to the plane (101) is 3.14 Å which is in well agreement with the expected value indicating the presence of stress-free conditions in the formation of these clusters[34,35,39].

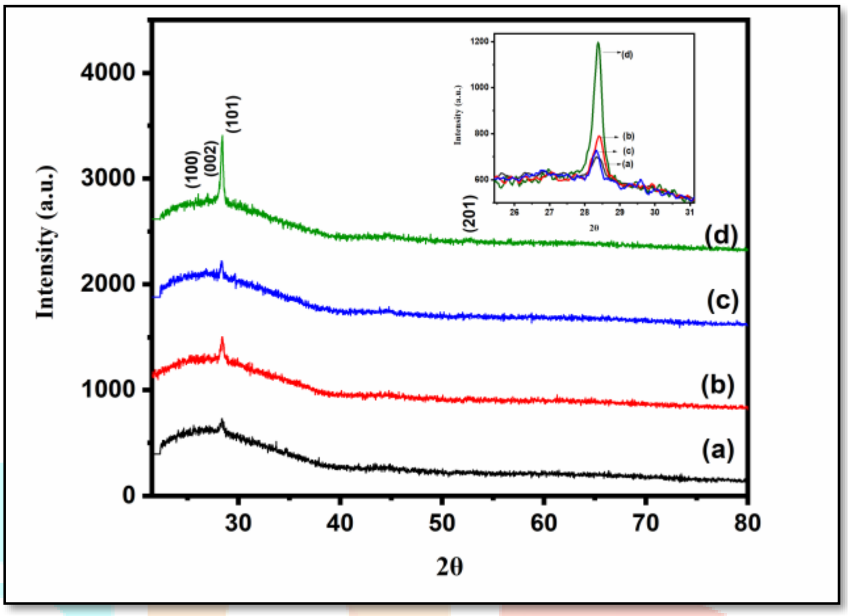

Figure 3: X-ray diffraction patterns of CdS for different deposition time (a) 0.5, (b) 1, (c) 1.5, and (d) 2 hours

The crystallite size of CdS thin films along reflection plane (101) was estimated using Debye Scherrer formula[36].
Where, λ is the wavelength of incident radiation (λ=1.54A˚), β is the full width at half maximum (FWHM) of the respective diffraction peak and θ is the Bragg diffraction angle.

Table 2Crystallite size of the CdS thin films prepared at different deposition time

Deposition time (hours)	2θ (degree)	β (radians)	Grain size (Å)
0.5	28.4	0.68	2.10
1	28.4	0.4	3.58
1.5	28.35	0.33	4.34
2	28.4	0.28	5.11

Table 2 illustrates the formation of CdS crystallites of nanometer size in all thin films for (101) plane and increase in size with deposition time, and thus exhibits improvement in the crystallinity of CdS hexagonal phase[19]. The deposition time significantly affect the formation of nano crystalline polycrystalCdS thin films. The decrease in the band gap and increase in the crystallite size with deposition time from 0.5 to 1.5 hours illustrates quantum confinement effect[37].

4. Conclusion:

Uniform, adherent CdS thin films were deposited on glass substrate using CBD method for different deposition time 0.5 to 2 hours in an interval of 0.5 hours. CdS thin films exhibited high optical transmittance in visible and NIR region of solar spectrum for deposition time 0.5 and 2 hours. The direct band gap was decreased from 2.46 to 2.1eV by increasing the deposition time from 0.5 to 1.5 hours and increased to 2.43eV with further increase in deposition time to 2 hours. XRD study confirmed the polycrystalline hexagonal phase of CdS thin films. The nanometer sized crystallites are formed in all CdS films and dominantly oriented along (101) plane. The decrease in band gap with increase in crystallie size showed quantum confinement effect.The entire study showcased CdS thin film deposited for 2 hours has high optical transmittance, high crystallinity and stable hexagonal phase formation with nanometer grains of CdS. Thin film CdS with high optical transmittance, wide range of optical band gap and stable hexagonal phase is preferred as a window layer in photovoltaic device and as a sensor in optoelectronic device.

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